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INTERFACE CONDUCTION
ELECTRON SPIN RELAXATION
IN LITHIUM-COPPER DOUBLE LAYERS

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INTERFACE CONDUCTION ELECTRON SPIN
RELAXATION IN LITHIUM-COPPER DOUBLE LAYERS

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ABSTRACT

Strong broadening of the conduction electron spin resonance spectra of double layers of lithium and copper was observed. The thickness of the lithium layers are of the order of 1μ while that of copper a few hundred angstroms. The broadening depends on the thickness of lithium but not on that of copper. The effect is attributed to spin relaxation at the lithium copper interface due to the difference in the spin-orbit constants of the metals.

РЕЗЮМЕ

Было обнаружено сильное расширение спектра парамагнитного резонанса электронов проводимости в двойных слоях лития и меди. Толщина слоев лития была порядка 1 микрона, а слоев меди - несколько сотен ангстрем. Расширение зависит от толщины слоев лития, но не зависит от толщины слоев меди. Это явление объясняется релаксацией спинов на общей поверхности двух слоев, которая обуславливается разницей в константах спин-орбитального взаимодействия этих двух металлов.

KIVONAT

Litium és rézből álló kettős rétegekben a vezetési elektron spin rezonancia spektrum erős kiszélesedését figyeltük meg. A litium rétegek vastagsága nagyságrendben 1μ , míg a réz rétegeké néhány száz angstrom. A kiszélesedés a litium réteg vastagságától függ, de a rézétől nem. Az effektust a két fém spin-pálya csatolási állandójának különbözőségéből eredő, az érintkezési felületen fellépő spin relaxációval magyarázzuk.

1. INTRODUCTION

The electron spin resonance /ESR/ spectrum of a metallic layer was first calculated by Dyson /1/. He calculated the surface impedance of samples under various conditions and included the effect of surface relaxation also. At the same time Fehér and Kip /2/ showed that for a more or less clean surfaced lithium the experimentally found spectra correspond well to a model where surface relaxation is neglected. Later experiments on very small lithium particles indicated also an extremely weak surface relaxation characterised by a surface collision spin flip probability of the order of 10^{-6} to 10^{-7} /3/. For clean surfaced aluminium and copper no surface relaxation was found /4//5/. Jánossy and Monod /5/ found in the transmission ESR spectra of aluminium foils coated with zinc a strong broadening over that of pure aluminium. The effect was attributed to relaxation due to the spin orbit interaction of zinc, strongly different from that of aluminium. It was demonstrated that the data could be fitted to a model where the additional relaxation is described by only one parameter independent of the thickness of the layer producing the relaxation. Nevertheless due to difficulties of sample preparation only rather thick zinc layers /0,5 to 0,9 μ / were fabricated so it could not be decided to what extent the relaxation comes from interface effects or relaxation of electrons diffusing into the bulk of the zinc layer.

In this paper we would like to demonstrate that large size dependent relaxation effects can be observed also by the conventional reflexion ESR technique by investigating relatively thick lithium films evaporated onto a second metal - in our case copper - producing the additional relaxation. We are able to show that in this system the additional relaxation is mainly due to a perturbation at the interface and not to bulk relaxation of the copper.

2. EXPERIMENTAL DETAILS

The lithium-copper layers were evaporated in a conventional vacuum system with a pressure during evaporation of 10^{-5} torr. The starting lithium

material was not very pure as seen from the bulk ESR linewidth of 15 Gauss. Lithium purifies during evaporation [2], lithium deposited onto glass or mica exhibited a linewidth of about 2 Gauss. ASARCO 99.999% high purity copper was used.

The double layers were fabricated by evaporating copper always first, the evaporation of lithium began after a few minutes. After the lithium evaporation the vacuum system was filled with air and the samples were quickly dipped into molten paraffin. Measuring the resistivity changes during the coating with paraffin we found that at most 0,1 μ lithium /on the free surface/ got oxidised. Although copper layers around 100 Å thick are usually not continuous we do not think this has any importance as the observed relaxation effect is proportional to the surface lithium covered by copper.

The method of preparing samples is rather crude but we shall compare only samples fabricated under the same evaporation run, i.e. under similar conditions - to arrive at our final conclusions. Samples with different lithium or copper thicknesses were obtained during the same evaporation by unshielding the substrates in two steps. Only two different kind of samples could be evaporated in one run.

The ESR measurements were done in a conventional spectrometer /JESME/ at X band and room temperature. The samples of about 1 cm² total surface were placed in the middle of a cylindrical cavity.

Four types of experiments were performed.

1. In the spectra of different thicknesses of lithium evaporated directly onto glass no variation of the relaxation rate was found for thicknesses ranging from 0,5 μ to 2 μ .
2. Lithium was evaporated at the same time onto glass and onto thin copper layers a few hundred Å thick. Although the results were not very reproducible we invariably found a much broader line for the lithium-copper double layers. In a typical case /fig. 1/ the broadening is 20 Gauss for a 2 μ thick lithium layer on copper.
3. Lithium of different thicknesses /1 and 2 μ / was evaporated onto copper substrates all having the same thickness / \approx 100 Å/. A strong variation of the linewidth is found, for double lithium thickness the resonance width is approximately half.
4. Lithium of the same thickness /1,7 μ / was evaporated onto copper layers of two different thicknesses /100 and 300 Å/. The spectra of the two kind of samples are identically broadened.

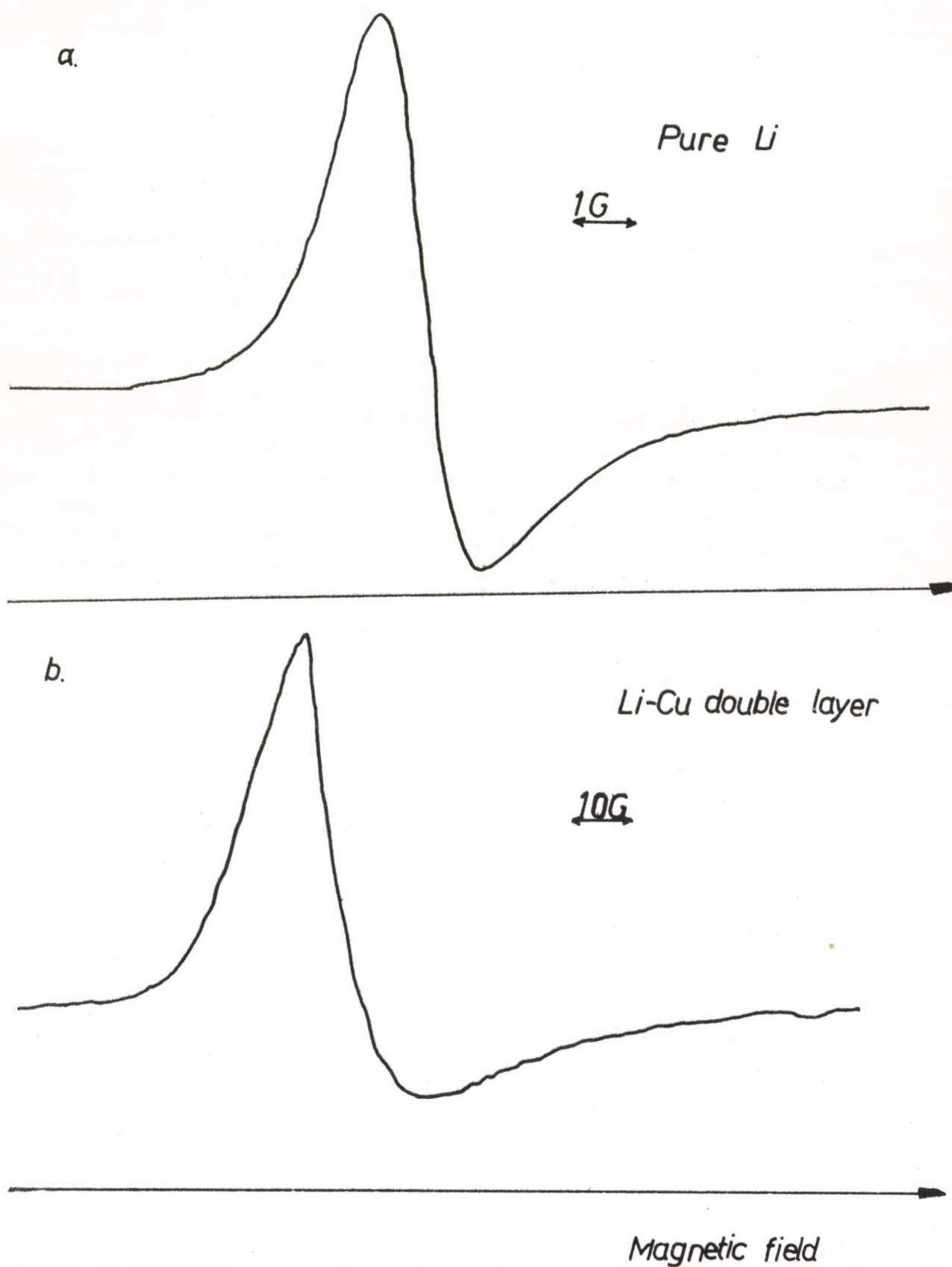


Fig. 1
ESR spectra of pure lithium and typical Li-Cu double layer. Lithium thickness $1,9\mu$, copper thickness 150\AA

3. DISCUSSION

The spin diffusion of a metal is well characterised by the spin penetration depth /1/ /6/:

$$\delta_{\text{eff}} = \left(\frac{2}{3} v_F \lambda T_1 \right)^{1/2}$$

where v_F is the Fermi velocity, λ is the electronic mean free path, T_1 the bulk conduction electron spin relaxation time. A polarised electron has a very high probability to become unpolarised when diffusing distances larger than δ_{eff} . This means also that in a double layer the magnetisation of the conduction electrons of one layer can be influenced only by that part of the second layer which lies within its spin penetration depth from the interface.

While it is not sure whether the zinc layer of reference /5/ was or was not thinner than its spin penetration depth, in the case investigated in this paper clearly both layers are an order of magnitude thinner than their δ_{eff} . Even a pessimistic estimation of the relaxation time of copper of 10^{-11} sec leads to $\delta_{\text{eff}}^{\text{Cu}} \approx 5000 \text{ \AA}$. From the measured relaxation time of lithium we estimate $\delta_{\text{eff}}^{\text{Li}} \approx 10 \text{ \mu}$. The copper layer is much thinner than its skin depth also; the lithium layer thickness is of the order of its skin depth /1,5 μ /. This latter condition would make a quantitative analysis difficult, qualitatively we believe that apart from changes in the form of the spectrum a model for a double layer with both layers thinner than the skin depth describes well the situation.

Menard and Walker /7/ pointed out that the resonance properties of a double layer interface can be characterised by three parameters. The first two describe relaxation at the interface in the same way as the surface relaxation parameter of Dyson /1/. The second parameter describes the reflectivity of the interface, i.e. the probability of an electron to be reflected from the interface without diffusing over. This parameter describes the coupling of the magnetisations of the layers and was introduced by Jánossy and Monod /8/ to explain the peculiar behaviour of the ESR of ferromagnetic-non-ferromagnetic double layers in a somewhat different way.

One may distinguish between two limiting cases depending on the strength of the coupling of magnetisations.

A. In strongly coupled systems /9/ the ESR spectrum has only one component. The width of the spectrum will be given by the average of the bulk and interface relaxations of the layers weighted by their thicknesses. The frequency is also a weighted average.

B. Decoupled systems. In general a two component spectrum appears,

the components differ from the spectra of the corresponding single layers only by the additional interface relaxation.

We believe that the experimental results are characteristic of a decoupled system where we observe the interface relaxation broadened resonance line of the lithium layer only. The copper resonance line is far too broad to be observed. /10/.

A copper thickness independent relaxation in the strong coupling limit could be expected only in case of a sufficiently slow bulk copper relaxation rate T_{Cu}^{-1}

$$T_{Cu}^{-1} \ll \frac{\ell_{Li} + \ell_{Cu}}{\ell_{Cu}} T_{obs}^{-1}$$

where ℓ_{Li} and ℓ_{Cu} are the layer thicknesses and T_{obs}^{-1} the observed relaxation rate. This would imply T_{Cu}^{-1} much less than $2 \cdot 10^{10} \text{ sec}^{-1}$, a rather improbable value in view of the results of Schultz and Latham /10/.

It is interesting to note that copper is insoluble in lithium to such an extent that Asik, Ball and Slichter /11/ did not observe any broadening of the ESR in lithium samples containing probably in a precipitated form about 1 % of copper. This is somewhat astonishing in view of our results as we would expect "surface relaxation" due to the precipitated particles also.

In conclusion we may state that the additional relaxation found in lithium-copper double layers is due to spin scattering at the interface and not to bulk relaxation in the copper. We have also shown that in the samples investigated the magnetisations of the layers are not coupled strongly.

The interface relaxation is most probably due to the change of the spin orbit coupling constant from one layer to the other, similarly to spin scattering on impurities in dilute alloys.

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